

Femtosecond laser nano-structuring of transparent materials

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New phenomena of anisotropic light scattering, anisotropic Cherenkov third-harmonic generation, photoinduced birefringence and anisotropic reflection from femtosecond direct-write structures are reviewed. The phenomena and recent direct observation reveal the smallest embedded structures ever created by light and new mechanism of light-matter interaction.

Keywords: laser machining, nano-structures, femtosecond, anisotropic, glass, self-organization

1. Introduction

The use of a femtosecond laser source to directly write structures deep within transparent media has recently attracted much attention due to its simplicity compared to lithographic methods, and its capability for writing in three-dimensions [1-3]. Tight focusing of the laser into the bulk of material causes non-linear absorption only within the focal volume, depositing energy that induces a permanent material modification [4-5]. A variety of photonic devices have already been created by translating a sample through the focus of a femtosecond laser [6-7]. Although molecular defects caused by such intense irradiation have been identified in fluorescence, ESR and other studies [8], the mechanism of induced modifications in glass is still not fully understood. New phenomena of light scattering [9-10] and Cherenkov third-harmonic [11] generation peaking in the plane of polarization during direct writing with ultrashort light pulses in glass have been reported. These observations were unexpected because the scattering of polarized light in the plane of light polarization in isotropic medium such as glass is always weaker compared to the orthogonal plane, since a dipole does not radiate in the direction of its axis. The phenomena were interpreted in terms of angular distribution of photoelectrons and sub-wavelength anisotropic index inhomogeneities. Another experiments demonstrated uniaxial birefringence of structures in fused silica written

by femtosecond light pulses [12]. The index change for light polarized along the direction of polarization of writing beam was much stronger than for the orthogonal polarization. The origin of this anisotropic phenomenon remained a mystery. Recently we observed a further anisotropic property in silica after being irradiated by a femtosecond laser – strong reflection from the modified region *occurring only along the direction of polarization of the writing laser* [13]. We show this can arise from a self-organized periodic sub-wavelength refractive index modulation. The femtosecond-laser-induced birefringence is therefore likely to be caused by these laterally-oriented small-period grating structures. Birefringence of this nature is well known as ‘form’ birefringence [14]. Our analysis suggests that this effect is also the primary cause of all anisotropic phenomena reported in the experiments on direct writing with ultrashort pulses in glass.

2. Anomalous anisotropic light scattering in glass

The laser radiation in Gaussian mode produced by regeneratively amplified mode-locked (120 fs pulse duration, 200 kHz repetition rate) Ti: sapphire laser operating at wavelength of 800 nm was used in the experiments [9-10]. Glass samples of ~3 mm thickness were placed on a stage under the optical microscope. The infrared laser radiation reflected by a dichroic mirror inside of the microscope was focused via a 20 X objective onto the sample. The pump spot size in the focus of the beam

spot size in the focus of the beam was 4.6 μm . Simultaneously, the irradiated spot was imaged in the visible spectral range via the microscope by using a colour CCD camera.

During the experiments on Ge-doped ($\text{GeO}_2 \sim 8 \text{ mol } \%$) silica glass strong blue luminescence (with a centre wavelength at 410 nm) of defect states (Ge-Si wrong bonds with the concentration of 10^{19} cm^{-3}) was observed. This luminescence (triplet luminescence) can be excited via the singlet-singlet transition by absorption of three pump photons or one UV photon of the third harmonic of the pump followed by quick nonradiative decay (with a decay time 1 ns) to the long-lived triplet level. When the pump (10 mW average power, 0.4 MW peak power, $2.5 \times 10^{12} \text{ W/cm}^2$ intensity in the focus of a beam) was focused slightly ($\sim 50 \mu\text{m}$) above the surface of the sample the shape of the spot of the blue luminescence imaged via the microscope and CCD camera was circular. Unexpectedly, it has been discovered that when the pump was focused inside the sample the spatial isotropy of the blue luminescence can be broken (Fig. 1): the luminescence scattering increases along the direction of the pump polarization, while the circular shape of the pump beam remains unchanged. If we rotate the di-

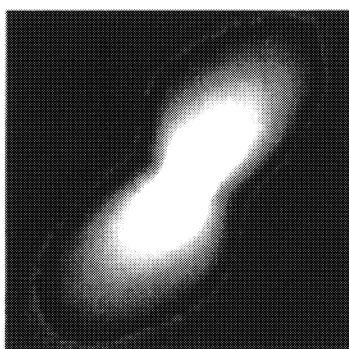


Fig. 1. Anisotropic blue luminescence in Ge-doped glass observed via microscope

rection of the pump polarization by using a half-wave plate, the elongated pattern of the blue luminescence follows this rotation. It should be noted that the blue luminescence was not polarized and self-focusing was not observed at peak powers used in the experiments. We called this phenomenon "propeller effect" due to the propeller-like shape of the luminescence spot in the focus of the pump beam. The observed phenomenon represents the first evidence of anisotropic light scattering which peaks in the plane of light polarization in isotropic media.

How can we explain this phenomenon? Firstly, let us estimate the size of a light spot which is produced by the isotropically emitted luminescence in the focal plane of the microscope objective. Assuming that the luminescence is excited by the three-photon absorption of the pump at

wavelength $\lambda = 800 \text{ nm}$ in a Gaussian beam with radius $r_0 = 2.3 \mu\text{m}$ or by the one-photon absorption of UV (267 nm) third harmonic of the pump and that it is emitted isotropically all along the length of a beam waist, the size of the light spot a can be estimated as: $a \approx \pi r_0^2 n / \lambda = 30 \mu\text{m}$, where $n = 1.45$ is the refractive index of silica glass. This estimate is in a good agreement with the transverse size of the blue propeller, which could be justified by ordinary (isotropic) luminescence. However, the longitudinal size of the blue propeller ($\sim 100 \mu\text{m}$) is about 4.5 times larger than the transverse size of the propeller. The fact that the blue luminescence is elongated along the pump polarization indicates that some additional momentum is acquired by the photons along this direction. We believe that such transformation of the momentum can be caused by the photoelectrons moving along the direction of pump polarization. Microscopic (much less than a wavelength of light) displacements of the photoelectrons along the direction of light polarization can lead to the *anisotropic* fluctuations of dielectric constant. Such fluctuations are obviously stronger along the direction of light polarization (in the direction of electron movement) compared to the perpendicular direction. The fluctuations of dielectric constant along the direction of light polarization induce index inhomogeneities which are elongated in the direction perpendicular to the pump polarization and which have $\mathbf{k}_w$ -vectors of spatial harmonics parallel to the direction of polarization. The anisotropic inhomogeneities scatter photons (e.g. the ultraviolet photons of the third harmonic of the pump) in the plane of light polarization. Considering the angle of scattering $\varphi = 80^\circ$ ($\tan \varphi = 3b/2z_0$, where $b = 100 \mu\text{m}$ is the longitudinal size of the "blue propeller", $2z_0 = 2\pi r_0^2 n / \lambda = 60 \mu\text{m}$ is the waist length of a pump beam), the size of these inhomogeneities can be estimated as: $d \approx \lambda_{uv} / (2n \sin \varphi) = 90 \text{ nm}$, where $\lambda_{uv} = 267 \text{ nm}$.

It should be pointed out that the scattering phenomenon described above must have strong wavelength dependence (λ^4), which is similar to the wavelength dependence of Rayleigh scattering of light. Rayleigh scattering is normally caused by isotropic density inhomogeneities and the anisotropy in the scattering (the scattering is stronger in the direction perpendicular to the light polarization) is explained by the fact that a dipole does not radiate along its axis. In contrast to Rayleigh scattering the anisotropy in the observed scattering is caused by the anisotropy of the inhomogeneities itself. The strong dependence of the scattering on the wavelength can explain the absence of noticeable changes in the shape of the infrared pump.

3. Anisotropic Cherenkov light generation in glass

In another experiment we used non-doped silica glass with weak absorption in the UV and a regeneratively amplified mode-locked Ti:Sapphire lasers operating at 1 kHz

repetition rate [11]. The laser radiation in Gaussian mode was focused via a lens with a focal distance of 6 cm into a fused silica (SiO_2) glass sample of 3 mm thickness. The pump spot size in the focus of the beam was about 14 μm . After passing through the sample the radiation was imaged on a screen of white paper.

When the pump (4 μJ energy, 4 mW average power, 33 MW peak power, $2.1 \times 10^{13} \text{ W/cm}^2$ intensity in the focus of a beam) was focused near the input surface or in the middle of the sample generation of a white light continuum was observed. When the radiation was focused closer to the output surface of the sample the generation of the white light continuum terminated. At that focus position a spectacular blue light pattern appeared on the screen and intensified (Fig.2). The pattern consisted of two crescent-like

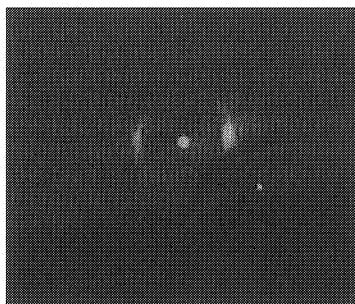


Fig.2. The pattern of ultraviolet light generated in a fused silica sample by intense linearly polarized infrared pump. The ultraviolet light is visualised on the screen via luminescence of the paper. Notice that the crescent-like lobes are located on both sides of the pump (the central bright spot of the pattern), along the polarization of the pump. The silhouette of a sample and a spot in the focus of the beam are on the lower right.

lobes on both sides of the pump, along the direction of light polarization. The intensity of light in the pattern increased over time and saturated after about 10 seconds of irradiation of the sample. A filter transmitting visible light placed between the sample and the screen completely blocked the blue pattern on the screen. This indicated that ultraviolet radiation generated in the sample is responsible for the observed pattern, which is visualised on the screen via luminescence of the paper in the blue spectral range. Analysing the spectrum of radiation from the output of the sample we confirmed the presence of 267 nm ultraviolet light, which is the third harmonic of the pump at 800 nm and which is generated via the third-order optical nonlinearity of silica glass. We measured an angle between the direction of propagation of the crescent-like ultraviolet light and the pump of about 21.6° . The Cherenkov angle of the third-harmonic propagation is estimated to be 20.9° using $n_{2 \text{ nm}} =$

1.499 and $n_{800 \text{ nm}} = 1.453$ for fused silica. This estimate is very close to the measured angle of propagation of the ultraviolet light, confirming that the crescents of light are generated via the Cherenkov mechanism of third-harmonic generation (THG). Two conclusions could be made on the basis of the experimental observations. First, the Cherenkov mechanism of the THG gives clear indication that longitudinal interfaces appear in the medium under intense irradiation. Secondly, the anisotropic distribution of the Cherenkov light with a maximum in the plane of pump polarization, in contrast to an isotropic "Cherenkov cone", indicates that the surface discontinuities appear only along the direction of polarization of intense light.

The observed phenomena represent unique optical effects in which information on light polarization is revealed macroscopically via enhanced light scattering and generation. The permanent changes induced by the polarized light in glass could be used for 3D polarization encoding and writing of photonic structures.

4. Anisotropic reflection from femtosecond-laser self-organized nanostructures in glass

More recently, we have observed a further anisotropic property in silica after being irradiated by a femtosecond laser – strong reflection from the modified region *occurring only along the direction of polarization of the writing laser* [13].

A regeneratively-amplified mode-locked Ti:Sapphire laser (150 fs pulse duration, 250 kHz repetition rate) operating at $\lambda = 850 \text{ nm}$ was used to directly write microstructures into a fused silica plate (40x40x1 mm). In the experimental arrangement, the collimated laser passes through an electronic shutter, variable neutral density filter and half-wave plate before a dichroic mirror reflecting only in the 400-700 nm region. The infrared laser light travels through the mirror and is focused through a 50x (NA=0.55) objective into the bulk of the sample, down to a beam waist diameter estimated to be $\sim 1.5 \mu\text{m}$. The silica sample is mounted on a computer-controlled linear-motor 3D translation stage (of 20 nm resolution). To simultaneously observe the writing process, a CCD camera with suitable filters and white light source is used.

A range of embedded diffraction gratings with overall dimensions $700 \mu\text{m} \times 700 \mu\text{m}$, each consisting of 100 rulings with 7 μm period were directly written towards the edges of the plate at a depth of 0.5 mm below the front surface. In every case, the speed of writing was 200 $\mu\text{m/s}$ and each grating ruling had only one pass of the laser. Pairs of embedded gratings were created with orthogonal writing polarizations directed parallel and perpendicular to the grating rulings respectively, with average fluence ranging from 270 mW ($\sim 1.1 \mu\text{J/pulse}$) down to 26 mW ($\sim 0.1 \mu\text{J/pulse}$). Additionally, pairs of embedded single lines of length 1 mm were written by the same method. Finally, a regular array of

40x40 'dots' with a pitch of 10 μm was directly written into the corner of the plate. Each 'dot' was produced by holding the sample translation stage stationary at each writing point, and irradiating for 3 ms (~ 750 pulses) using the electronic shutter.

After writing, the samples were viewed through the silica plate's polished edges using a 200x microscope incorporating a color CCD camera. During inspection, the structures were illuminated with a randomly-polarized white light source in a direction along the viewing axis, either from below the structure (opposite side to microscope objective), or above the structure (through the microscope objective). The embedded structures were examined through the edge nearest to them, and the array of dots was examined in two

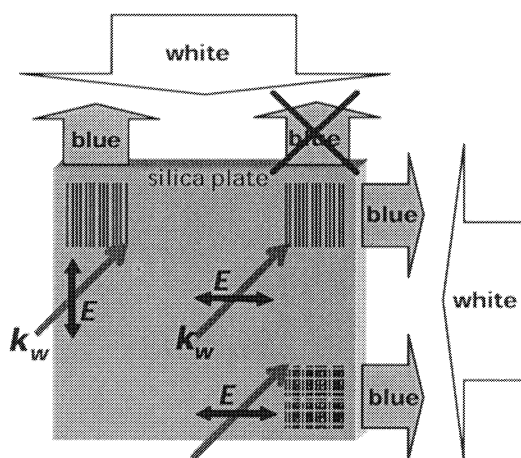


Fig. 3. Schematic showing the anisotropic reflection from embedded photonic structures. Reflection only occurs for incident light parallel to the electric-field vector of the incident writing laser. The magnified region (bottom) illustrates the laser-induced self-organized periodic nanostructuring responsible.

orthogonal directions. A striking reflection was observed in the blue spectral region from a number of the structures when illuminated via the viewing objective. Closer analysis revealed that the reflection *only* occurred when the viewing axis was both parallel to the electric field vector of the writing beam and the structure was written with pulse energy greater than $\sim 0.5 \mu\text{J}$. This indicates that the observed reflectivity is both fluence dependent and highly anisotropic. Fluorescence cannot account for the observation due to the directional dependence. Fig. 3 shows a schematic of the reflection phenomenon. As can be seen, the macroscopic shape of the photonic structures does not determine the direction of the anisotropic reflection.

Figure 4 shows microscope images of the reflection from several directly-written embedded structures. The illuminating light in all cases was incident above the samples through the viewing objective and set to a level that ensured

that the weak-contrast microstructure itself was not imaged. The spatial position of the embedded objects relative to the focus of the microscope objective was checked beforehand by illuminating from below, when the embedded structure in all cases could be clearly observed. The displayed images were chosen from regions of modification created with a pulse energy of $\sim 0.9 \mu\text{J}$. In each example the orientation of the direct-write laser's electric-field is indicated, while the k_w -vector marks the incident direction of the writing laser beam. Fig. 4(a) displays the reflection from a single line which has dimensions $\sim 1.5 \mu\text{m}$ into the page due to the focal width of the beam, and $\sim 30 \mu\text{m}$ down the page due to the beam's confocal parameter, enhanced by self-focusing effects. Figure 4(b) shows the reflection from the side of a 100-line grating, with its rulings going into the depth of the page. Not all of the 100 lines of the grating contribute to the recorded reflection because of the $\sim 2 \mu\text{m}$ focal depth of the imaging objective. Nevertheless, the reflection is considerably enhanced compared to the single line in Fig. 4(b). Fig. 4(a, b) also show the result of imaging an identical structures, except written with orthogonal polarization. From this orientation, no reflecting structure at all can be observed. Fig. 4(c) shows a section of the 40x40 array of

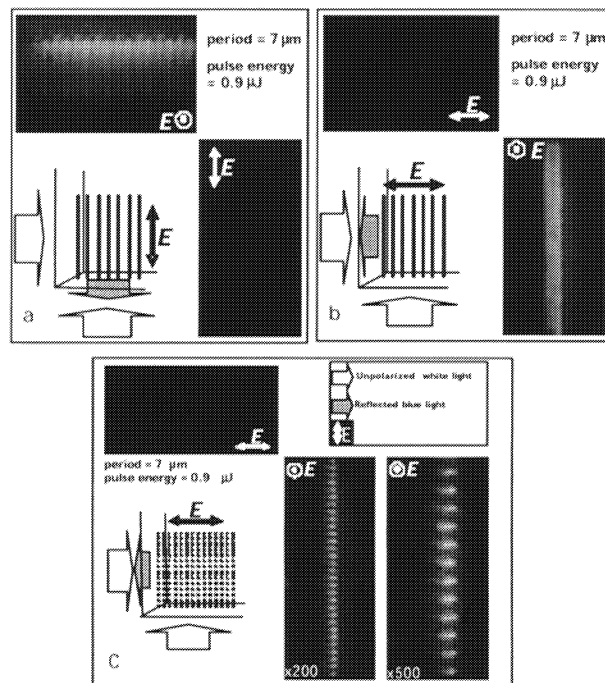


Fig. 4. CCD camera images of the reflection from different embedded structures directly-written with the laser. (a) Reflection from 100 lines one behind another into the page, (b) reflection from single line, (c) array of 'dots' which show reflectivity dependent only on the writing polarization orientation.

'dots' described above, once again producing strong reflection

tion along the writing beam polarization axis. These particular structures are interesting because they are approximately circular with a diameter of $\sim 1.5 \mu\text{m}$ when viewed from the direction of the writing laser, and therefore have a uniform cross section. However when viewed from a direction orthogonal to the axis of the writing beam's polarization, there is no reflecting component as Fig. 4(c) clearly demonstrates. The reflected light observed from the structure shown in Figure 4(b) is analyzed yielding the spectrum displayed in Fig. 5. This data shows a strong peak at 460 nm, which accounts for the blue color when observed under a microscope.

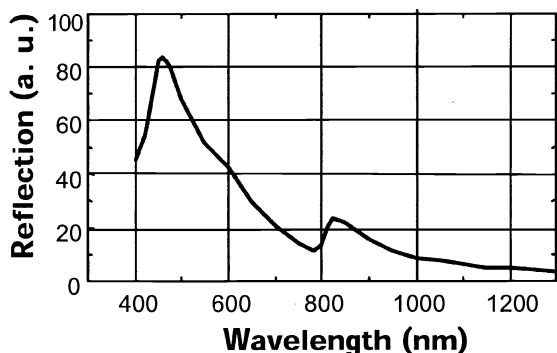


Fig. 5 Spectrum of the reflected light from the structure shown in Fig. 4(b).

We suggest that the anisotropic reflectivity can only be explained as a consequence of Bragg reflection from a self-organized periodic structure. Indeed, a modulation in refractive index of period $\sim 150\text{nm}$, produced only along the direction of the incident laser's electric field can account for the observed anisotropic reflection at $\lambda \sim 460\text{nm}$ ($\lambda/2n$). Such a grating does not reflect when viewed edge on. The magnified section at the bottom right-hand corner of Fig. 2 demonstrates how the periodic nanostructuring is arranged in the case of a single $1.5\mu\text{m}$ diameter 'dot'. The orientation and the period of the nanostructure are almost identical to the periodic nanostructure implicated in the phenomenon of anisotropic light scattering [9]. Closer inspection of Fig. 5 shows an additional smaller peak at 835nm . This suggests that an extra grating component may be formed which has double the periodicity of the laser-induced structures. Alternatively, this long-wavelength reflection could be a second-order diffraction peak of a grating component with period equal to the wavelength of the incident light. Surface ripples with a period equal to the wavelength of incident laser radiation and that are likewise aligned in a direction orthogonal to the electric field have been observed in experiments involving laser deposition.¹⁵ Nevertheless, we believe that our data is the first reported evidence of periodic nanostructures (much

smaller than the wavelength of incident light) being generated within the bulk of a material. We speculate they arise from a mechanism associated with the creation of a hot electron plasma by multiphoton absorption of incident light. Anisotropic index inhomogeneities are then induced by electrons moving along the direction of light polarization [9]. Self-organized nanostructures are in turn produced by a pattern of interference between the incident laser radiation and a plasmon-polariton wave generated within the sample. Positive feedback leads to exponential growth of the periodic nanostructures in the plane of light polarization, which become frozen within the material.

Further microscope characterization was carried out on the same samples by positioning the fused silica plate between crossed polarizers and illuminating from below along the direction of the original writing beam. With the polarizers oriented at angles of $\pi/4$ to the writing-beam electric-field vector, this enables regions of birefringence to be identified. The onset of birefringence occurs at a writing fluence level $\sim 0.5 \mu\text{J/pulse}$, equal to that found in the case of the anisotropic reflection. This strongly suggests that the mechanism responsible for inducing reflection along the writing beam polarization axis is the same mechanism that causes birefringence in the orthogonal direction. The femtosecond-laser-induced birefringence, which up to now has not been well understood, is therefore likely to be caused by the laterally-oriented small-period grating structures. Birefringence of this nature is well known as 'form' birefringence [14].

5. Direct observation of self-organised nanostructures in glass

In our experiments [16] we used commercially available synthetic silica glass, ED-H (Tosoh Quartz Corp., brands of wet silica with OH ~ 50 ppm) of $10\text{ mm} \times 10\text{ mm} \times 2\text{ mm}$ size. The laser radiation in Gaussian mode produced by regenerative amplified mode-locked Ti:Sapphire laser (150 fs pulse duration, 200 kHz repetition rate) operating at a wavelength of 800 nm was focused via $100\times$ (NA=0.95) microscope objective into the silica glass samples placed on the XYZ piezo-translation stage. The beam was focused at $\sim 100 \mu\text{m}$ below the surface and the beam waist diameter was estimated to be $\sim 1 \mu\text{m}$.

After laser irradiation the sample was polished to the depth of the beam waist location. The surface of the polished sample was analyzed by scanning electron microscope (SEM, JEO, model JSM-6700F) and Auger electron spectroscopy (AES, PHI, model SAM-680). Secondary electron (SE) images and backscattering electron (BE) images of the same surface were compared (Fig. 6). It is well known that the SE image reveals the surface morphology of a sample, while the BE image is sensitive to the atomic weight of the elements or the density of material constituting the observa-

tion surface. The SE images of the polished silica sample indicate that the morphology of an irradiated sample in the examined cross-section does not change, namely, a void does not exist. On the other hand, the BE images reveal a

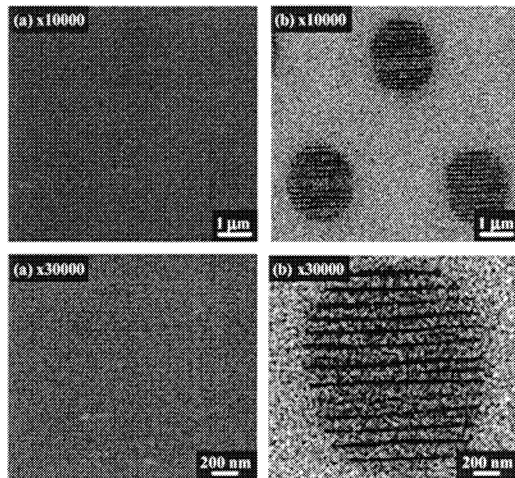


Fig.6 (a) Secondary electron images of silica glass surface polished close to the depth of focal spot. (b) Light "fingerprints": Backscattering electron images of the same surface. The magnification of the upper and lower images is $\times 10000$ and $\times 30000$ respectively.

periodic structure of stripe-like dark regions with low density of material and of ~ 20 nm width which are aligned perpendicular to the writing laser polarization direction. We speculated, based on the fact that the elements constituting the sample are silicon and oxygen (average molecular weight of SiO_2 glass ~ 60.1), that the oxygen defects were formed in the regions corresponding to dark domains of the BE image, which reduce the average molecular weight in these regions ($\text{SiO}_{2-x} \sim 60.1-16x$). To test this suggestion we carried out Auger spectra mapping of silicon and oxygen on the same surface by Auger electron spectroscopy. The Auger spectra signal of the oxygen in the regions corresponding to dark domains in the BE image is lower compared to other regions, indicating low oxygen concentration in these domains. Furthermore, there is some indication that the intensity of the oxygen signal is stronger in the regions between the dark domains of the BE image. On the other hand, the intensity of the silicon signal is the same in the whole imaged region. These results indicate that the oxygen defects (SiO_{2-x}) are periodically distributed in the focal spot of the irradiated region. The Auger signal intensity is proportional to the concentration of element constituting the surface, which gives an estimate to the value $x \sim 0.4$.

We observed the decrease of the gratings period with an increase of the exposure time. The grating periods were about 240 nm, 180 nm and 140 nm for the number of light pulses of 5×10^4 , 20×10^4 and 80×10^4 respectively and for the pulse energy of 1 μJ . This indicated a logarithmic de-

pendence of the grating period Λ on the number of light pulses N_{pulse} . The dependence of the observed periodic nanostructures on pulse energy for a fixed exposure time was also investigated and an increase of the period with the pulse energy was observed. Grating periods of 180 nm, 240 nm and 320 nm were measured at pulse energies of 1 μJ , 2 μJ and 2.8 μJ respectively and for the number of light pulses of 20×10^4 (Fig. 7a).

6. Mechanism of embedded self-organised nanostructures in glass

The following explanation of the observed phenomenon is proposed [16]. The light intensity in the focus of the beam is of 10^{14} W/cm^2 , which is high enough for multiphoton ionization of glass matrix. Once a high free electron density is produced by multiphoton ionization the material has the properties of plasma and will absorb the laser energy via one-photon absorption mechanism of inverse Bremsstrahlung (Joule) heating. The light absorption in the electron plasma will excite bulk electron plasma waves (Langmuir wave). These are longitudinal waves with the electric field component parallel to the direction of propagation. Such electron plasma wave could couple with the incident light wave only if it propagates in the plane of light polarization. Initial coupling is produced by inhomogeneities induced by electrons moving in the plane of light polarization. The coupling is increased by a periodic structure created via a pattern of interference between the incident light field and the electric field of the bulk electron plasma wave, resulting in the periodic modulation of the electron plasma concentration and the structural changes in glass. Positive feedback will lead to exponential growth of the periodic structures oriented perpendicular to the light polarization, which become frozen within the material. The electron plasma wave is efficiently generated only with wave vector k_{pl} ($k_{\text{pl}} = \omega_{\text{pl}}/v_{\text{pl}}$, where v_{pl} is the speed and ω_{pl} is the angular frequency of the plasma wave) in the plane of light polarization and only in the direction defined by conservation of longitudinal component of the momentum (insert Fig. 7b). The latter condition is similar to the condition in Cherenkov's mechanism of nonlinear wave generation. The period of the grating is defined by this momentum conservation condition: $k_{\text{gr}} = 2\pi/\Lambda = k_{\text{pl}}^2 - k_{\text{ph}}^2$, where $k_{\text{ph}} = n/c$ is the wave vector, n is the refractive index and c is speed of light. This relation can also be used to estimate the value of electron plasma wave vector $k_{\text{pl}} = [k_{\text{ph}}^2 + (2\pi/\Lambda)^2]^{1/2}$ which gives $k_{\text{pl}} = 43.5 \times 10^4 \text{ cm}^{-1}$, assuming $\omega_{\text{pl}} = 2.36 \times 10^{15} \text{ s}^{-1}$ and $\Lambda = 240 \text{ nm}$. The dispersion relation for the electron acoustic waves or Langmuir waves is as follows: $\omega_{\text{pl}}^2 = \omega_p^2 + 3/2 (v_e^2 k_{\text{pl}}^2)$, where $\omega_p = (N_e e^2 / \epsilon_0 m_e)^{1/2}$ is the plasma frequency, N_e is the electron density, m_e is the electron mass, e is the electron

charge, $v_e = (2k_B T_e / m_e)^{1/2}$ is the thermal speed of electrons, T_e is the electron temperature, k_B is Boltzmann constant. Taking into account the energy conservation condition $\omega_{pl} = \omega$ the momentum conservation relation and the above dispersion relation it is possible to calculate the dependences of the grating period on the electron temperature and the electron plasma density (Fig. 7a, b). Assuming that the electron temperature is proportional to the pulse energy due to one-photon absorption of light by the electron plasma, the experimental dependence of the grating period on the pulse energy is in very good agreement with the theoretical prediction (Fig. 7a). The theoretical estimates give high electron

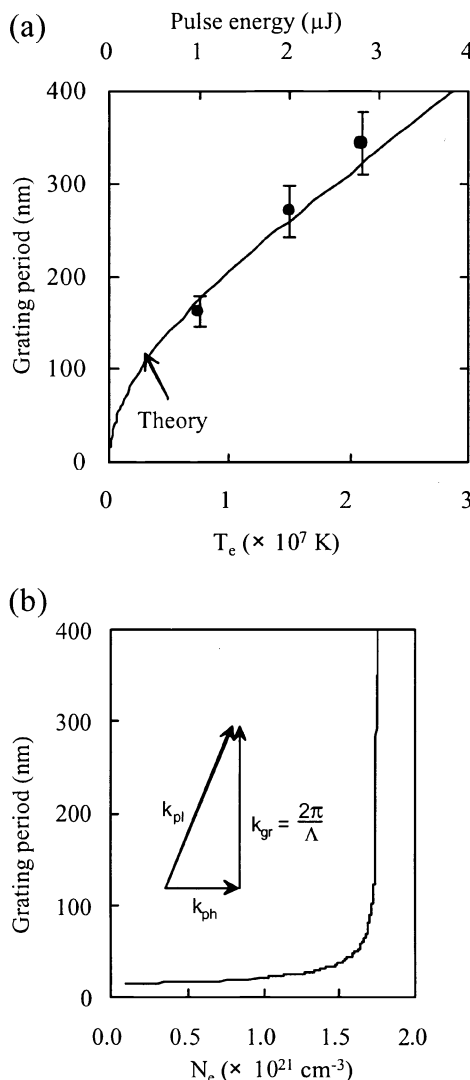


Fig.7 Theoretical dependence of self-organized grating period on electron temperature for $n_e = 1.7 \times 10^{21}$ cm $^{-3}$ (a) and electron concentration for $T_e = 2 \times 10^6$ K (b). Experimental grating periods versus pulse energy are also shown (a). Insert is a wave vector phase-matching diagram (b).

concentrations of 1.7×10^{21} cm $^{-3}$ and high electron temperatures of 2×10^6 K for the interpretation of the experimental results.

Based on the above mechanism of grating formation it is possible to give the following explanation of the observed formation of stripe-like regions with low oxygen concentration. The interference between the light wave and the electron acoustic wave will lead to modulation of the electron plasma concentration. The plasma electrons are created in the process of breaking of Si-O-Si bonds via multi-photon absorption of light which is accompanied by the generation of a Si-Si bonds, non-bridging oxygen-hole centers (NBOHC, = Si-O $^{\cdot}$) and interstitial oxygen atoms (O_i). Such oxygen atoms are mobile and can diffuse from the regions of high concentration. Negatively charged oxygen ions can be also repelled from the regions of high electron concentration. Small thickness of these regions compared to the period of the grating could be explained by highly nonlinear dependence of light absorption on the electron concentration. In addition, accumulated structural changes in glass during irradiation result in reduction of the electron plasma concentration. Therefore, the period of the grating decreases with the increase of number of writing pulses (Fig. 7). It should be pointed out that the irradiated regions in glass reflected light only in the direction parallel to the polarization of the writing laser which has been also observed in other experiments. Moreover, the periods of the gratings in our experiments are very close to 150 nm which is the period of gratings responsible for the reflection and scattering phenomena.

Apart from the fundamental importance of the observed phenomenon as the first evidence of interference between light and electron acoustic waves, the observed light "fingerprints" are the smallest embedded structures ever created by light. The related phenomena of anisotropic light scattering, reflection and birefringence has been observed in various glasses and crystals. This indicates that the reported light-induced nanostructuring is a universal phenomenon in transparent materials and could be useful for optical recording and photonic crystal fabrication.

References

- [1] K.M. Davis, K. Miura, N. Sugimoto, and K. Hirao, *Opt. Lett.* **21**, 1729 (1996).
- [2] K. Miura, J.Qiu, H. Inouye, T. Mitsuyu, and K. Hirao, *Appl. Phys. Lett.* **71**, 3329 (1997).
- [3] E.N. Glezer and E. Mazur, *Appl. Phys. Lett.* **71**, 882 (1997).
- [4] M.D. Perry, B.C. Stuart, P.S. Banks, D. Feit, V. Yanovsky, and A.M. Rubenchick, *J. Appl. Phys.* **85**, 6803 (1999).
- [5] K. Yamada, T. Toma, W. Watanabe, K. Itoh, and J. Nishii, *Opt. Lett.* **26**, 19 (2001).
- [6] Y. Kondo, K. Nouchi, T. Mitsuyu, M. Watanabe, P.G. Kazansky, and K. Hirao, *Opt. Lett.* **24**, 646 (1999).

- [7] D. Homoelle, S. Wielandy, A. L. Gaeta, N.F. Borrelli, and C. Smith, *Opt. Lett.* **24**, 1311 (1999).
- [8] H. Sun, S. Juodkasis, M. Watanabe, S. Matsuo, H. Misawa, J. Nishii, *J. Phys. Chem. B* **104**, 3450 (2000).
- [9] P.G. Kazansky, H. Inouye, T. Mitsuyu, K. Miura, J. Qiu, K. Hirao, and F. Starrost, *Phys. Rev. Lett.* **82**, 2199 (1999).
- [10] J. Qiu, P. G. Kazansky, J. Si, K. Miura, T. Mitsuyu, K. Hirao, and A. Gaeta, *Appl. Phys. Lett.* **77**, 1940 (2000).
- [11] P.G. Kazansky, H. Inouye, T. Mitsui, J. Qiu, K. Hirao, F. Starrost, "Anisotropic Cherenkov light generation by intense ultrashort light pulses in glass," in *Quantum Electronics and Laser Science*, (Optical Society of America, Washington, D.C., 2000), paper QFA3.
- [12] L. Sudrie, M. Franko, B. Prade, A. Mysyrowicz, *Opt. Commun.* **171**, 279 (1999).
- [13] J. Mills, P.G. Kazansky, E. Bricchi, J. Baumberg, *Appl. Phys. Lett.* **81**, 196 (2002).
- [14] M. Born and E. Wolf, *Principles of Optics* (Cambridge University Press, UK, 1999), p. 837
- [15] S.R.J. Brueck and D.J. Ehrlich, *Phys. Rev. Lett.* **48**, 1678 (1982).
- [16] G. Kazansky, Y. Shimotsuma, J. Qiu and K. Hirao, "Nanostructuring of transparent materials by ultrashort light pulses," CLEO, Baltimore, 2003, postdeadline paper CThPDD.